

Crystal-melt \pm fluid equilibria versus late-stage fluid - rock interaction in granitoid rocks of the South Mountain Batholith, Nova Scotia: whole rock geochemistry and oxygen isotope evidence

Daniel J. Kontak, David F. Strong et Robert Kerrich

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[Aller au sommaire du numéro](#)

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Résumé de l'article

On a examiné la dévotion de la chimie et des isotopes de l'oxygène dans le Batholite péralurain de South Mountain (daté à 370 Ma) en Nouvelle-Écosse. Le diarsisme des suites magmatiques, tant tardives que précoces, est uniforme en ce qui a trait à la plupart des éléments majeurs et en traces, y compris les terres rares (T.R.). Cette continuité du chimisme met en lumière le rôle prépondérant joué par les équilibres bain-cristal durant le fractionnement progressif de ce bain via la série granodiorite-rhyolite-leucogranite (groupe I). En revanche, les leucogranites du groupe II, associés aux venues pegmatitiques dans la région de New Ross, ont des signatures chimiques (plus particulièrement des spectres de terres rares extrêmement appauvris en T.R. légères et montrant des profils concaves pour les T.R. lourdes avec des anomalies positives à Gd-Dy) qui mettent en évidence le rôle majeur joué par les équilibres fluide-phase dans l'évolution de ces roches.

Les compositions isotopiques de l'oxygène sur roche totale sont uniformes à $10,6 \pm 0,5$ ‰ (K-14) peu importe la proximité des contacts, le degré de fractionnement ou la présence d'altérations deutériques tardives. On attribue la variation observée (environ 1,8 ‰) à une hétérogénéité primaire de la source et non pas aux processus de fractionnement des cristaux. Les minéraux séparés des pegmatites montrent des déviations par rapport à l'équilibre magmatique basé sur le fractionnement des quartz et feldspaths, ce qui semble trahir un ré-équilibre du feldspath avec des fluides de température faible issus du magma (6 ‰ calculé du fluide est 7-10 ‰). Par contre, un des greisens examinés met en lumière une interaction avec des fluides enrichis en O (0 ± 0 ‰ calculé 11,5-14 ‰ pour T*300-400 °C) dérivés probablement de l'encaissement métasédimentaire (i.e., le Groupe de Hegama). Une faible valeur de δ (3,6 ‰) en provenance d'un seul échantillon sur roche totale indique que les eaux météoriques ont aussi joué un rôle, quoique mineur, dans l'évolution du batholite.

CRYSTAL-MELT+FLUID PHASE EQUILIBRIA VERSUS LATE-STAGE FLUID-ROCK
INTERACTION IN GRANITOID ROCKS OF THE SOUTH MOUNTAIN
BATHOLITH, NOVA SCOTIA: WHOLE ROCK GEOCHEMISTRY
AND OXYGEN ISOTOPE EVIDENCE

Daniel J. Kontak*, David F. Strong
Department of Earth Sciences, Memorial University of Newfoundland
St. John's, Newfoundland A1B 3X5

and

Robert Kerrich
Department of Geological Sciences, University of Saskatchewan
Saskatoon, Saskatchewan S7N 0W0

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The chemical and oxygen isotopic evolution of the 370 Ma, peraluminous South Mountain Batholith of Nova Scotia has been examined. The chemistry of early- to late-stage magmatic suites is uniform with respect to most major and trace elements, including the rare earth elements (REE). The continuity of the chemistry reflects the dominant role of melt-crystal equilibria during progressive fractionation of the melt through the sequence granodiorite-monzogranite-leucogranite (group I). However, leucogranites (group II) associated with pegmatite occurrences in the New Ross area have chemical signatures, especially their REE patterns (extreme LREE depletions and concave HREE profiles peaking at Gd-Dy), which indicate that fluid-phase equilibria played an important role in the evolution of these rocks.

Whole-rock oxygen-isotope compositions are uniform at 10.6 ± 0.5 ‰ (N-14) regardless of proximity to contacts, degree of fractionation or presence of late-stage deuteric alteration. The observed variation (ca. 1.8 ‰) is attributed to primary heterogeneity in the source region, and not crystal fractionation processes. Mineral separates from pegmatites indicate deviations from magmatic equilibrium based on quartz-feldspar fractionation. This is interpreted to reflect re-equilibration of feldspar with low temperature fluids of magmatic derivation (calculated $\delta^{18}\text{O}$ of the fluid is $7-10$ ‰). In contrast, one of the greisens examined records interaction with ^{18}O enriched fluids (calculated $\delta^{18}\text{O}$ composition of 11.5-14 ‰ for T=300-400°C), which were probably derived from the enveloping meta-sedimentary country rock (i.e., Meguma Group). A low ^{18}O value (3.6 ‰) for one whole-rock sample indicates that meteoric water also played a role, albeit minor, in the evolution of the batholith.

On a examiné l'évolution de la chimie et des isotopes de l'oxygène dans le Batholite péralumineux de South Mountain (daté à 370 Ma) en Nouvelle-Ecosse. Le chimisme des suites magmatiques, tant tardives que précoces, est uniforme en ce qui a trait à la plupart des éléments majeurs et en traces, y comprises les terres rares (T.R.). Cette continuité du chimisme met en lumière le rôle prépondérant joué par les équilibres bain-cristal durant le fractionnement progressif de ce bain via la série granodiorite-monzogranite-leucogranite (groupe I). En revanche, les leucogranites du groupe II, associés aux venues pegmatitiques dans la région de New Ross, ont des signatures chimiques (plus particulièrement des spectres de terres rares extrêmement appauvris en T.R. légères et montrant des profils concaves pour les T.R. lourdes avec des anomalies positives à Gd-Dy) qui mettent en évidence le rôle majeur joué par les équilibres fluide-phase dans l'évolution de ces roches.

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[Traduit par le journal]

INTRODUCTION

The evolution of granitoid bodies involves early magmatic processes dominated by melt-crystal equilibria, which is succeeded by late-stage magmatic and post-magmatic processes in which the role of a fluid phase becomes more important as the solubility of aqueous fluids is reduced in residual melts and saturation occurs (e.g., Burnham, 1979).

*Present address: Nova Scotia Department of Mines and Energy, P.O. Box 1087, Halifax, Nova Scotia B3J 2X1)

The latter stage is manifested most prominently in the formation of pegmatites, metasomatic zones and areas of mineralization, particularly the granitophile association (e.g., Sn, W, U, Li, Be: see Strong (1981) for a general review). Because of the inevitable involvement of at least some fluid phase of variable origin (e.g., juvenile, metamorphic, meteoric) during the evolutionary history of granitoid bodies, and consequently some modification of magmatic mineralogy and chemistry, it is not always easy to discern the origin of alteration and mineralization in granites (e.g., Stone and Exley, 1986; Manning and Exley, 1984;

Kontak and Corey, 1988). It is essential that the features characteristic of these different processes be recognized as they provide the foundation for determining the nature and origin of granitoid rocks and related mineral deposits (e.g., see recent papers in Taylor and Strong (1985) and HHPG (1985)).

To examine this problem, we have focused our study on a suite of rocks from the eastern part of the South Mountain Batholith (SMB), where the extreme end-members, as well as the transitional stage, of these processes are well represented. For example, early granodiorites, representing the most primitive component of the SMB, and late-stage monzogranites, leucomonzogranites and pegmatites, representing the most evolved members, all outcrop in the eastern part of the SMB. While the magmatic and late hydrothermal processes operative within the SMB have been examined by numerous workers (e.g., McKenzie and Clarke, 1975; Logothetis, 1984; Chatterjee and Strong, 1984; Ford and O'Reilly, 1985) and their chemical and petrographic signatures "defined" (e.g., extreme modification of rare-earth element (REE) patterns in greisen zones: Chatterjee and Strong, 1984; Strong and Chatterjee, 1985), the transitional stage between these two extremes remains poorly understood. For example, during the latest stage of crystallization and earliest post-magmatic stage when volatiles become increasingly more important there will be a re-equilibration of whole rock mineralogy and chemistry due to the influence of the fluid phase (e.g., Ford and O'Reilly, 1985). It is this transitional stage which is the focus of the present study as we try to address the problem of distinguishing the boundary between crystal-melt ± fluid versus rock-fluid equilibria.

We first discuss the petrography and chemical features of the granitic rocks in order to establish magmatic versus orthomagmatic or hydrothermal trends. The oxygen isotopic signatures of whole rocks and mineral separates are reported, and the results are then discussed in the context of magmatic versus hydrothermal processes and integrated with other petrological data.

REGIONAL GEOLOGICAL SETTING AND EVOLUTION OF THE SOUTH MOUNTAIN BATHOLITH

The SMB is a ca. 370 Ma, post-tectonic, peraluminous granitoid batholith which intruded Cambro-Ordovician metasedimentary rocks of the Meguma Group. Level of emplacement is considered to have been 1-3 kb (i.e., 4-12 km) and the chronological sequence of intrusion was granodiorite, monzogranite and leucomonzogranite. Pegmatites and mineralized leucogranite centres are locally present within the SMB. These field relationships indicate, therefore, that the formation of late-stage, fluid-rich systems was indeed an important and integral part of the evolution of this batholithic complex. Much of the late-stage fluid activity was centred in and around the New Ross area, the geology of which is summarized by O'Reilly *et al.* (1982).

Previous workers have concluded from various lines of evidence that the SMB, although strongly peraluminous in nature, did not originate by anatexis of the Meguma Group (Clarke and Muecke, 1985, and references therein). Instead, another

crustal source material is considered the likely parent, with a minor contribution of Meguma Group lithologies having occurred during the magmatic (Clarke and Halliday, 1980) and post-magmatic stages (Kubilius, 1983). While crystal fractionation processes have generally been considered responsible for the chemical evolution of the SMB (McKenzie and Clarke, 1975; Smith *et al.*, 1986), it has been suggested that late-stage fluid-melt-crystal equilibria were also important in controlling the distribution of at least some trace elements in the more evolved members of the SMB. For example, Clarke and Muecke (1981) have attributed the patterns and abundances of the REE in the leucomonzogranites to be partly a result of removal via an exsolved mobile fluid. However, the recent identification of accessory mineral phases such as monazite and xenotime (M.A. MacDonald, personal communication, 1987) can also account for the general depletion of the LREE and MREE in the SMB (Smith *et al.*, 1986) and, thus, the necessity of a fluid phase to account for this feature, as Clarke and Muecke (1981) advocated, is perhaps not warranted.

SAMPLE LOCATIONS AND PETROGRAPHY

Samples were selected from the eastern part of the SMB within an area that has recently been remapped (MacDonald *et al.*, 1987). The sample localities and simplified geology of the region are shown in Figure 1. Because of the high density of pegmatite bodies and related mineralization in the New Ross area, our study was focused here. However, material was also selected from other localities in the eastern part of the SMB for comparative purposes. In addition, samples from both close to and well removed from contacts with metasedimentary rocks were also collected in order to evaluate the possibility of contamination in response to wall-rock assimilation.

No granodiorites were sampled as Longstaffe *et al.* (1980) and Chatterjee *et al.* (1985) provided several analyses of this lithology which is remarkably uniform in terms of its oxygen isotopic composition ($\delta^{18}\text{O} = 10.1\text{-}10.8\text{‰}$, N=13). Monzogranites with highly variable proportions of biotite, muscovite and cordierite are represented by samples which are both proximal (No. 14: Turner tin) and distal (Nos. 9, 31, 53) to mineralized centres. Late-stage, medium- to fine-grained leucomonzogranites containing variable proportions of biotite and muscovite, also represent both barren (Nos. 17-1, 17-2, 39) and mineralized (Nos. 11-2, 20, 21, 26-1, 26-2, 33, 35, 37A) areas. Samples of pegmatites formed at different stages of the evolution of the batholith were also taken. For example, the pegmatite from the Pockwock Lake area (No. 49) represents a small, local segregation within a biotite-muscovite-garnet monzogranite. In contrast, the Keddy's (Nos. 18, 19), Reeve's (No. 22C), Morley's (No. 38) and Grassy Brook (No. 25) pegmatite occurrences represent more evolved pegmatoids (based on the geochemistry of alkali feldspars, unpublished data of Kontak) which are associated with mineralized leucomonzogranites. The Long Lake (Nos. 34-1, 34A) and Walker moly (No. 11) pegmatites are associated with coarse quartz-K-feldspar intergrowths, have banded aplite-pegmatite units, and are richly mineralized. A more detailed

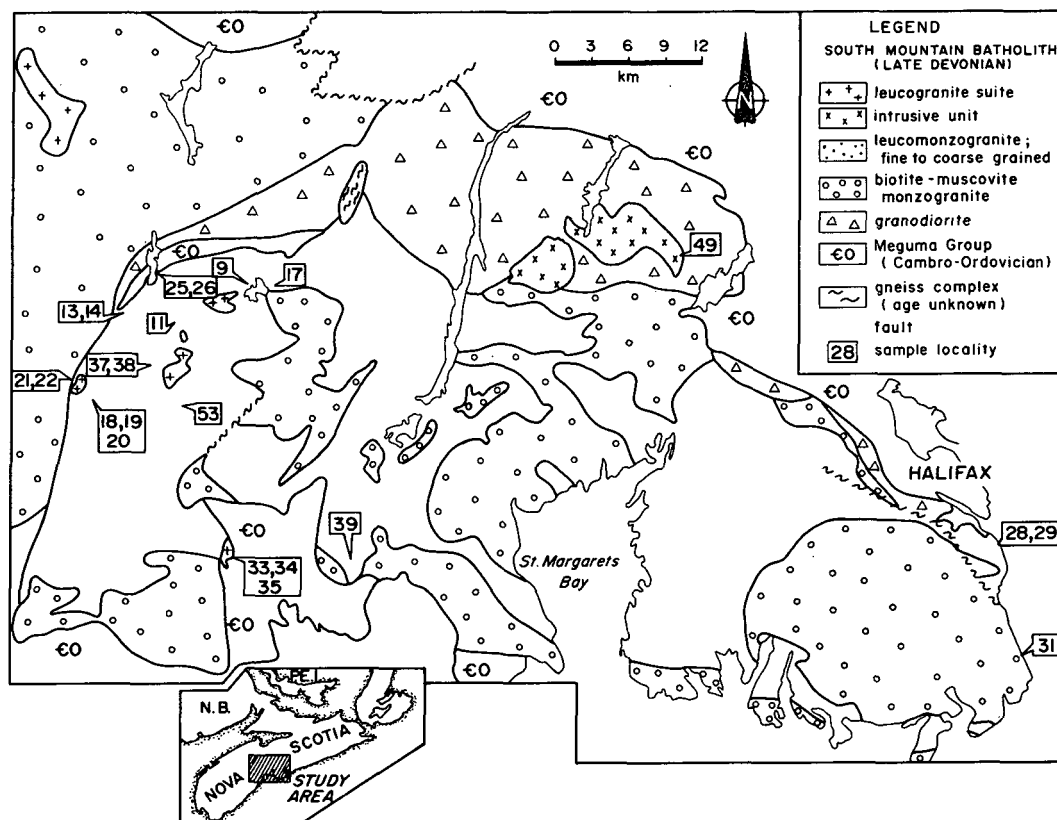


Fig. 1. Simplified geological map of the eastern part of the South Mountain Batholith (modified after MacDonald *et al.*, 1987) showing sample localities.

account of these localities is provided by O'Reilly *et al.* (1982). Two zones of intense greisenization within a biotite-muscovite monzogranite from the southeastern part of the SMB are represented by samples 28 and 29.

The petrographic features of all the units discussed herein are described in detail by numerous previous workers (e.g., Charest, 1976; McKenzie, 1974; Logothetis, 1984), so only the salient aspects are discussed below.

The monzogranites are medium- to coarse-grained, have biotite > muscovite, contain coarse K-feldspar megacrysts and may contain cordierite (e.g., No. 31). In thin section biotites characteristically contain abundant inclusions, the K-feldspars (generally monoclinic variety) may contain relict plagioclase (i.e., from replacement), muscovite is of several textural varieties (see Ham and Kontak, this volume), plagioclase (An_{20-25}) is generally fresh, and K-feldspar is variably perthitic.

The leucogranites, fine- to medium-grained and texturally variable, are subdivided into two groups based on petrographic and chemical features (note also that group II leucogranites are associated with pegmatites). The most obvious difference between the groups is the presence of 1-2% biotite in the former group (note that in samples 33 and 35 chlorite is considered to have formed after biotite), whereas it is absent in group II and andalusite is an important constituent. Group I leucogranites contrast with the monzogranites in having fewer inclusions in biotite, which is also more commonly chloritized. In addition, plagioclase

is more albitic in composition (An_{0-10}), zoning is rarely developed, and inclusions of secondary sericite and apatite are common. The K-feldspar rarely contains perthite lamellae, and finer grained phases are typically microcline. In group II leucogranites many of the above features are also present, with the following additional observations: (i) plagioclase is mostly pure albite and contains numerous inclusions of secondary sericite and apatite; (ii) muscovite is more abundant and its texture ragged; and (iii) andalusite may be quite abundant. The andalusite, of subhedral to anhedral shape, occurs as both isolated grains and also with mantles of muscovite. It is difficult to establish from textural criteria alone whether the andalusite is of magmatic or post-magmatic origin (cf. Clarke *et al.*, 1976).

We emphasize that the above features are broad generalizations, but they serve to illustrate that important differences are readily apparent. Perhaps the most significant of these differences is that the more evolved rocks appear to have equilibrated with a fluid phase which is indicated by the presence of albite, alteration products in plagioclase, presence of microcline, texturally variable muscovite, chloritization of biotite and the abundance of probably secondary andalusite.

ANALYTICAL METHODS

Sixteen whole rocks were analyzed for major and trace elements and REE (Tables 1 and 2). Major element analyses were obtained at Memorial

WHOLE-ROCK CHEMISTRY RESULTS

Major Elements

The granites are all enriched in silica with 73.1 to 76.7 wt.% SiO_2 . With respect to the cationic elements, only Mg appears to define a clear pattern with decreasing values from the monzogranites through the leucogranite suites. For Ca and Fe there is no clear trend which we attribute to the following:

(1) Fe, although most consistently enriched in the monzogranites, is elevated in other rocks due to the presence of minor sulphides and celadonic muscovite.

(2) Ca is highly variable in the leucogranite and overlaps the values for the monzogranites (0.36 to 0.52 wt.%). Much higher values of Ca in some of the leucogranites (up to 0.72 wt.%) are due to the presence of fluorite and apatite which are inferred to be of secondary origin based on textural evidence.

Both Mn and Ti have low abundances (near or below detection limits) in all of the suites. There is general decrease in the $\text{K}_2\text{O}:\text{Na}_2\text{O}$ ratio from the monzogranites through to the leucogranite suites. This is commensurate with the change in plagioclase to more albitic composition.

Levels of phosphorous are similar in all suites, except for sporadic enrichment in a few cases where apatite is present, as mentioned above. There is also a change in the peraluminous index of the suites from the monzogranites ($x=1.14$), leucogranites I ($x=1.12$) to leucogranites II ($x=1.26$). In the latter suite, samples with the higher A/CNK values (Nos. 11-2, 26-2) contain abundant andalusite; however, their lower total alkali contents suggest that some post-magmatic

changes in the bulk composition of the samples may have occurred.

Trace Elements

The salient aspects of the trace element data, excluding the REE's, are summarized as follows:

F, B, Li: the volatile elements are generally all low (cf. Tischendorf (1977) for typical abundance levels in various granitoid rocks). The highest F values (ca. 3000 ppm) are found in the leucogranites, but these contrast, for example, with F contents of about 1-2 wt.% ($x=0.78 \pm 0.36$, N=26) in leucogranites hosting the East Kemptville tin deposit (Kontak, 1987).

Rb, Sr, Ba: these elements show covariations typical of those expected from crystal fractionation processes (i.e., Rb increase; Sr and Ba decrease) operative in granitoid systems (cf. McCarthy and Hasty, 1976). The low Ba and Sr contents in the leucogranites are perhaps worthy of comment. It is not possible to say whether these low values are due to real depletions as a result of extended differentiation or, alternatively, if the albitization of plagioclase may have caused liberation of the elements as the albite structure does not incorporate Sr or Ba.

K/Rb ratios of the three suites are quite distinctive (Fig. 2), and for the more evolved leucogranites the values correspond to the pneumatolytic field of Shaw (1968).

Cs, Ga, Nb, Ta: this group of incompatible elements shows general enrichment throughout the sequence of monzogranites to leucogranites, although Cs is erratically distributed compared to the other elements.

Zr, Hf: Zr shows a gradual depletion with progressive evolution of the suites and Hf is

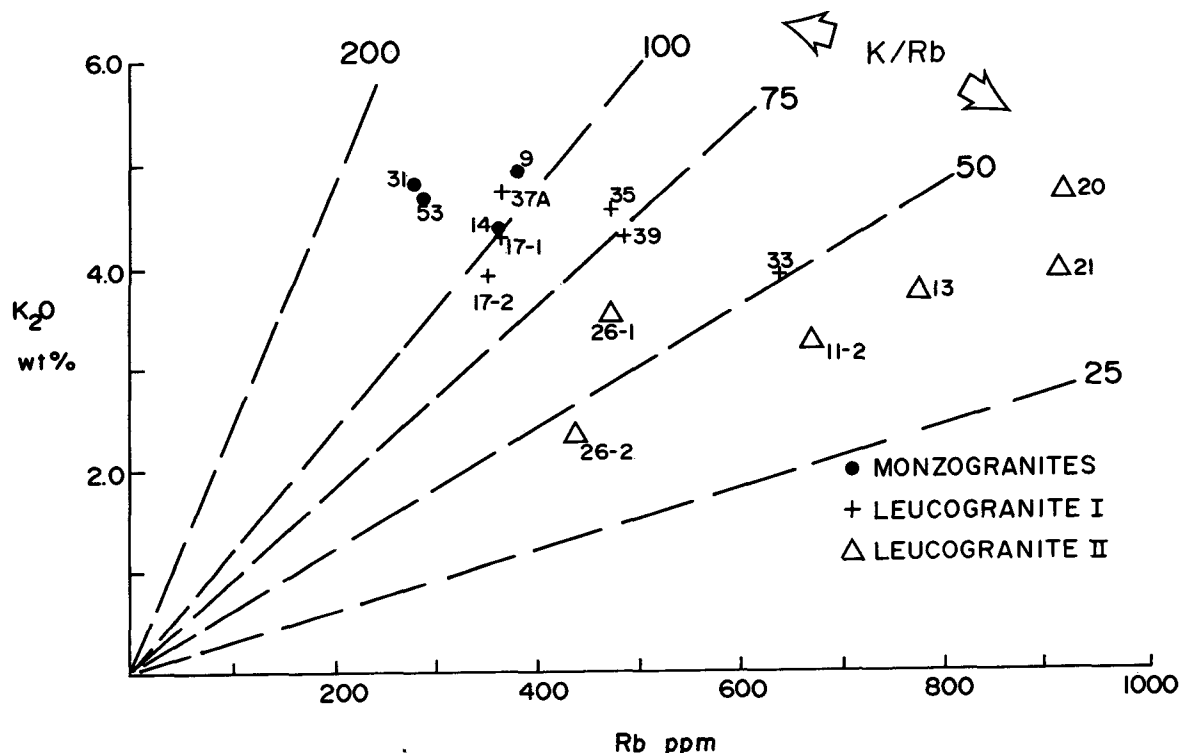


Fig. 2. K versus Rb plot for granites of the South Mountain Batholith.

consistently low; accordingly, the Zr/Hf ratio diminishes.

U-Th: these two elements generally behave systematically throughout differentiation in the SMB (e.g., Chatterjee and Muecke, 1982), but no consistent trends are apparent in these particular rocks (Fig. 3). Th values in the monzogranites ($x=7.4 \pm 1.5$) are enhanced compared to the other rocks which generally have <4 ppm Th, but U behaves erratically and appears to have been mobile suggesting, perhaps, U mobility during a late hydrothermal overprinting event.

Sn-W: both Sn and W are elevated above contents considered typical of magmatic values in low Ca granitoid rocks in general (Turekian and Wedepohl, 1961), with Sn more anomalous than W. The anomalously high Sn content of 60 ppm for sample 14 is related to the proximity (i.e., in direct contact) of a late stage, fine-grained dyke (No. 13) which itself contains 100 ppm Sn.

Zr vs Nb; Ta vs Zr; Zr vs Rb diagrams: in this series of variation diagrams (Fig. 4) the different groupings of granitoid rocks are clearly distinguished. The general trends are interpreted to reflect magmatic processes, except for the Zr versus Rb plot where a certain amount of the Rb enrichment is probably due to orthomagmatic or even later hydrothermal activity. These diagrams clearly illustrate the progressive evolution of the groupings toward relatively more differentiated compositions with higher Ta and Nb abundances and lower Zr contents, and illustrate the retention of certain geochemical signatures diagnostic of the magmatic crystal fractionation of minerals such as zircon.

Rare Earth Elements: Chondrite-normalized REE patterns for all the rocks are presented in Figure 5. The monzogranites are characterized by LREE enrichment ($La_N = 40-112$), strongly fractionated patterns ($La_N/Yb_N = 9-33$), and moderately negative Eu anomalies ($Eu/Eu^* = 0.2-0.3$); all features that have been recognized in other metaluminous to peraluminous granitoid suites. The patterns and absolute abundances (total 10 REE = 110 ± 53 ppm) are similar to those of monzogranites elsewhere in the SMB (Clarke and Muecke, 1981) and also to those of the Musquodoboit Batholith (MacDonald and Clarke, 1986).

Group I leucogranites have LREE's slightly depleted compared to the monzogranites ($La_N = 10-30$), moderately fractionated patterns ($La_N/Yb_N = 5-22$), and moderate to extreme Eu depletion ($Eu/Eu^* = 0.08-0.46$ to 0.01 ; note that some values are below detection limit (see Table 1)). The absolute abundances of the REE's (total 10 REE = 38 ± 13 ppm) are also lower than those for the monzogranites.

Group II leucogranites are characterized by a further absolute depletion of the REE's (total 10 REE = 12 ± 4 ppm), generally flat chondrite-normalized profiles ($La_N/Yb_N = 0.5-9.2$) with a slight concave shape to the HREE portion. These patterns and abundances are similar to leucomonzogranites of the SMB reported in Clarke and Muecke (1981) and have been documented from the evolved phases of other granitic complexes (e.g., Nabalek 1986; Noyes *et al.*, 1983).

In a plot of total REE versus K/Rb (Fig. 6), the different granitic suites are clearly distinguished, and such a plot illustrates well

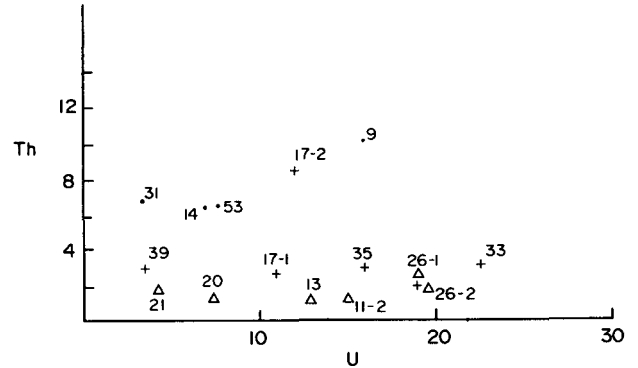


Fig. 3. Th versus U plot for granites of the South Mountain Batholith. Symbols as in Figure 2.

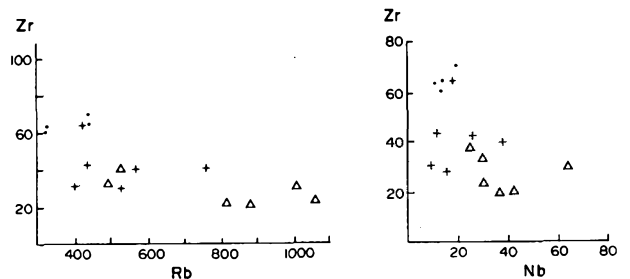
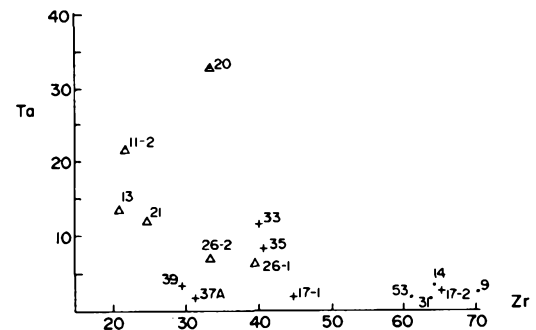


Fig. 4. Ta versus Zr, Zr versus Rb, and Zr versus Nb plots for granites of the South Mountain Batholith. Symbols as in Figure 2.

the trend of progressive depletion of the REE's as the rocks become geochemically more evolved.

Comparing, therefore, the REE patterns for these three suites we note the following prominent features: (1) an overall depletion of the total REE's, due mainly to the decrease in the LREE abundances as Yb_N values are similar for all the suites; (2) a marked increase in the magnitude of the negative Eu anomaly within the leucogranite suites; and (3) a tendency for the HREE profile for the leucogranites (particularly Group II) to be concave.

OXYGEN ISOTOPE RESULTS

Whole Rock Results

Oxygen isotope data for 15 whole rock samples have a narrow range of $\delta^{18}O$ values, regardless of distribution (i.e., proximity to granite-sediment

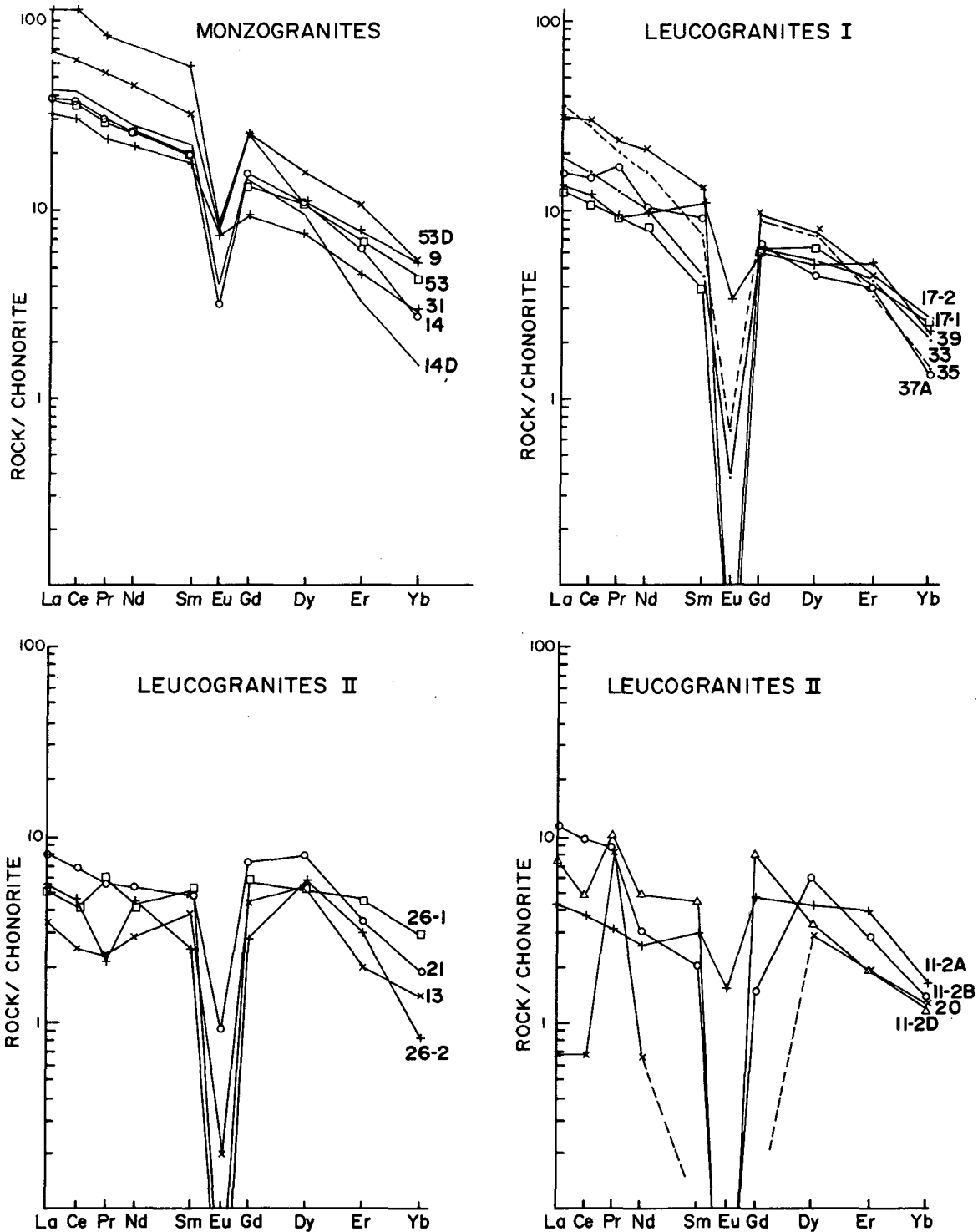


Fig. 5. Chondrite-normalized REE plots for granites of the South Mountain Batholith.

contact), with a total range of between 9.5 to 11.3 ‰ (mean of 10.6 ± 0.5 ‰). There is no systematic variation of values corresponding to the groupings discussed above, nor with respect to geochemical indices of differentiation (e.g., SiO_2 in Fig. 7). The only deviation from this homogeneity is an anomalously low value of 3.6 ‰ obtained for the fine-grained dyke rock from the

Turner tin prospect. A value in this range necessitates the involvement of a component of low ^{18}O water at some time in the post-magmatic history of this rock (e.g., Taylor, 1978).

The whole-rock data are similar to the results obtained by Longstaffe *et al.* (1980) and Chatterjee *et al.* (1985a) for the SMB granites and are also similar to data from other peraluminous, crustal-

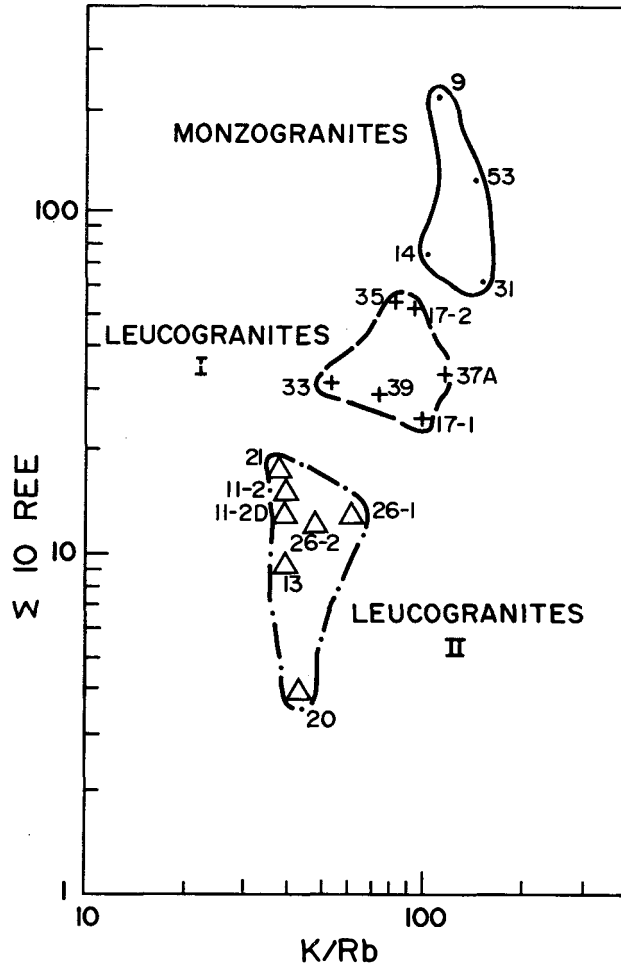


Fig. 6. K/Rb versus total REE plot for granites of the South Mountain Batholith. Symbols as in Figure 2.

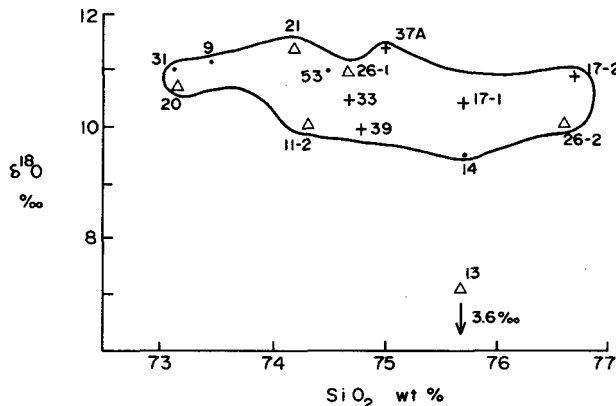


Fig. 7. Whole rock $\delta^{18}\text{O}$ values versus wt.% SiO_2 plot for granites of the South Mountain Batholith. Symbols as in Figure 2.

derived suites throughout the world (see Sheppard, 1986) that have values of 10–14 ‰.

Results for Mineral Separates

Data for quartz, K-feldspar and muscovite are plotted in Figures 8 and 9. There is a large variation for minerals between the different

localities despite the similarities of whole rock chemistry and oxygen isotope values referred to above. For example, among the pegmatitic occurrences the $\delta^{18}\text{O}$ values for K-feldspar are much more variable (8.9 to 11.6 ‰) than the corresponding whole rock values (10–11.3 ‰). In contrast, quartz, which is much more retentive of its primary $\delta^{18}\text{O}$ value, shows a significantly smaller range of 10.4–11.6 ‰, with the only exception being a value of 9.9 ‰ for quartz from Walker moly. The two muscovites from the pegmatites gave similar results of 8.2 and 8.8 ‰.

Compared to the mineral data from Longstaffe *et al.* (1980), our data are similar for quartz and muscovite, but K-feldspar contents are much more variable (8.9–11.6 ‰ versus 10.4–11.3 ‰).

Two examples of greisen developed within a biotite-muscovite monzogranite (No. 31) form an isotopically distinct population. Both quartz (18.2 ‰) and muscovite (12.0 ‰) from sample 28 are anomalously enriched in ^{18}O by ~6 ‰ compared to the data for other pegmatites presented herein and granites in general. Similarly, the K-feldspar from a greisenized monzogranite (No. 29) is also enriched, with a $\delta^{18}\text{O}$ value of 12.2 ‰.

For magmatic systems under conditions of isotopic equilibrium, quartz-K-feldspar and quartz-muscovite fractionations are 1 ± 0.5 ‰ and 2.2–2.5 ‰, respectively (Taylor and Epstein, 1963; Taylor, 1968, 1978). Of the three quartz-muscovite pairs analyzed, only sample 49 would appear to have retained magmatic equilibrium, and for the seven quartz-K-feldspar pairs only one (No. 38) has retained fractionations within the magmatic field. A plot of the quartz-feldspar data in Figure 9 further illustrates this point and indicates that for five samples the feldspars have been ^{18}O enriched, while one sample has been ^{18}O depleted.

Also shown in Figure 9 are the data from Longstaffe *et al.* (1980). All of their samples correspond to the magmatic field and indicate that equilibrium has been maintained in the samples they studied.

DISCUSSION

The chemical and isotopic data presented for the granitic rocks are interpreted to represent a relatively simple evolution which can be accommodated by dominantly fractional crystallization processes \pm late aqueous fluid phase equilibria with a certain degree (the real extent cannot be demonstrated with the current data base) of late chemical modification due to hydrothermal activity. This case is best argued using elements such as Nb, Ta, Zr and the REE's which appear to have retained dominantly magmatic trends and patterns despite petrographic evidence indicating sporadic late-stage modification of the leucogranites by hydrothermal fluids. However, the enrichment of the leucogranites in some elements (e.g., Rb) and potential mobility of others (e.g., U, Th) indicate that fluid-rock interaction was probably responsible for some, albeit not a great deal of, chemical change. Relevant to the study of felsic rocks in general, therefore, is the identification of the following parameters:

(1) determination of the boundary between magmatic (crystal-silicate melt \pm aqueous fluid

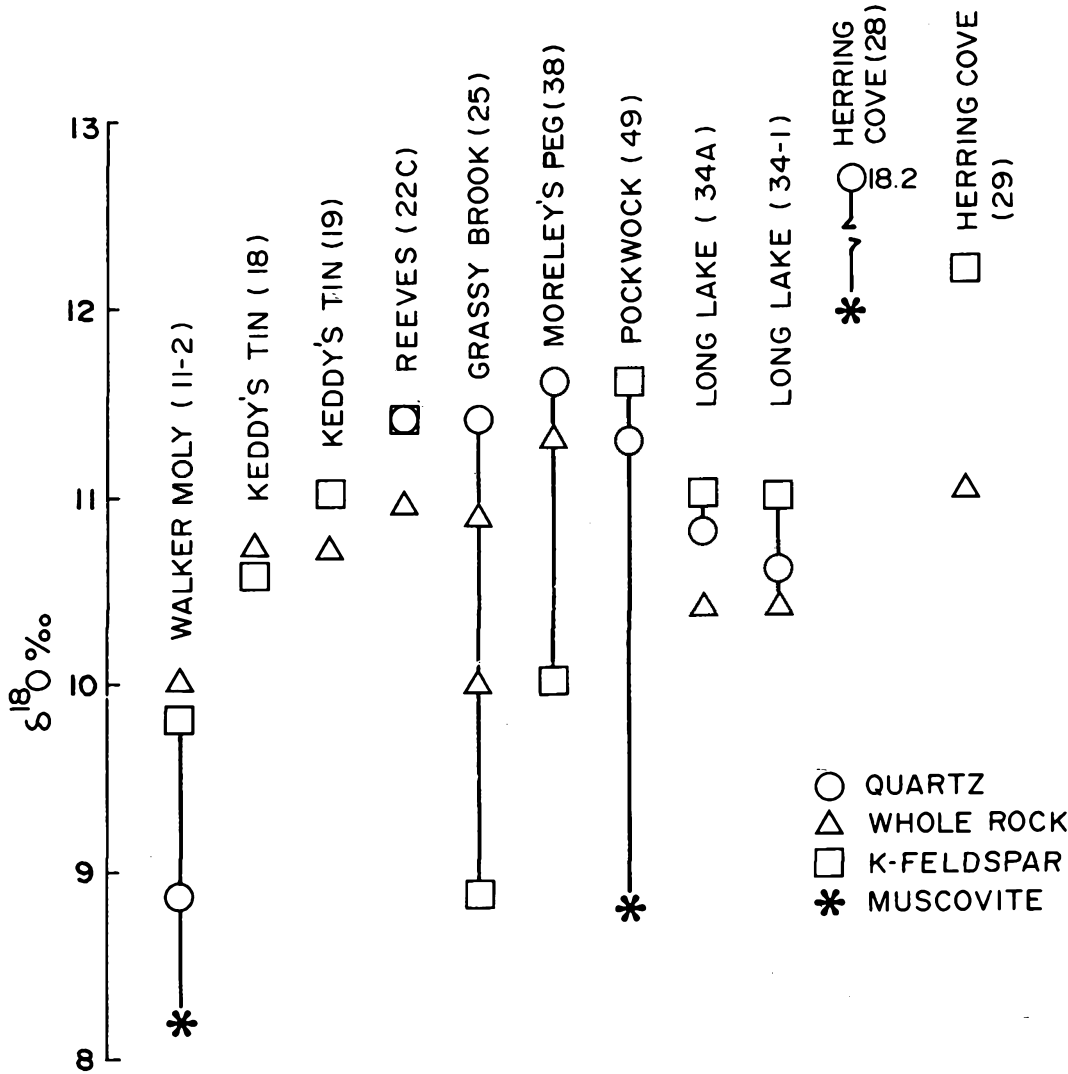


Fig. 8. $\delta^{18}O$ values for mineral separates from pegmatites and greisens in the South Mountain Batholith.

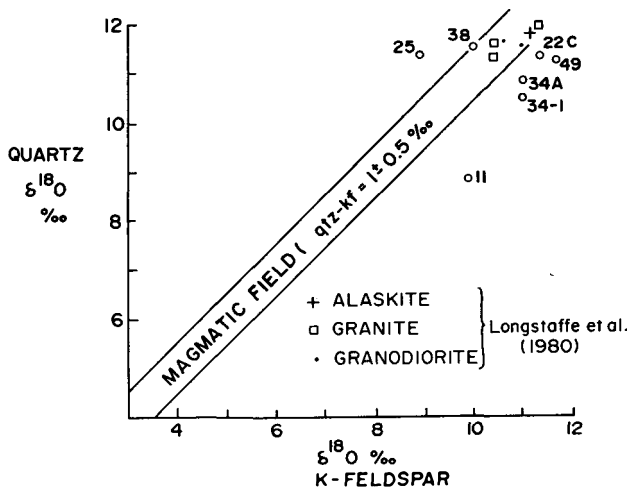


Fig. 9. $\delta^{18}O$ quartz versus $\delta^{18}O$ feldspar plot for mineral separates from pegmatites and greisens in the South Mountain Batholith. The magmatic trend represents quartz-feldspar fractionations calculated and measured for undisturbed plutonic igneous rocks.

equilibria) and post-magmatic (fluid-rock equilibria) concentrations for elements that are particularly susceptible to hydrothermal enrichment and depletions;

(2) in the case where a late fluid phase has been active, the nature of the source reservoir (i.e., magmatic, meteoric, metamorphic). This, of course, has direct relevance to the origin of ore deposits.

Magmatic Versus Post-Magmatic Processes

The data presented herein for the granitic rocks define consistent trends for many elements, including those which are considered to be relatively immobile (e.g., Pearce *et al.*, 1984), despite the presence of essentially cryptic alteration which characterizes some of the geochemically more evolved rocks. Although we do not suggest that all the rocks owe their origin to a common liquid line of descent, it does appear as if broadly comparable processes have been operative throughout a large part of the SMB (i.e., common magmatic histories). In order to examine more

closely the role of crystal-silicate melt versus fluid-rock \pm melt equilibria we will confine ourselves in this discussion to the REE profiles and abundances. However, we note that examination of the covariation of Ba versus Sr and Rb on log-log plots using our data and previously published data for the SMB indicate that fractional crystallization was the dominant petrological process during the evolution of the SMB (cf. McCarthy and Hasty, 1976; Cocherie, 1986).

The progressive decrease in the abundances of the REE (Figs. 5, 6) is due mainly to the LREE, a feature commonly observed in granitoid rocks or felsic systems in general (Miller and Mittlefehldt, 1982, 1984). Three potentially important processes are considered and evaluated, viz., liquid-state diffusion, aqueous fluid phase complexing, and crystal fractionation.

Liquid-state diffusion has been considered as a possible mechanism to explain the enhanced concentration of LILE and the decrease of LREE in high-silica rhyolites (Hildreth, 1979, 1981) and granite systems (Ludington, 1981; Michael, 1984; Tuach *et al.*, 1986). The petrological features of the SMB do not resemble such systems and, therefore, it is unlikely that such processes played a very dominant role in the chemical evolution of this batholith.

Flynn and Burnham (1978) demonstrated experimentally that the LREE can be complexed by a Cl-bearing aqueous fluid phase and Taylor and Fryer (1982, 1983) and Taylor *et al.* (1981), among others, applied these results to natural situations. In the case of the SMB there is little field or petrographic evidence to suggest that a fluid phase was important during the progression from granodioritic to monzogranitic suites. Thus, it is unlikely that the LREE patterns are related to stripping due to Cl-complexes via expulsion of an aqueous fluid phase from the melt during fractionation. However, the leucogranite suites may represent a different situation which we discuss in further detail below.

The ability of crystal fractionation processes to account for the progressive depletion of LREE in felsic systems is supported by the high partition coefficients that have been measured for accessory mineral phases such as monazite, xenotime, allanite, sphene and apatite (e.g., Gromet and Silver, 1983; Sawka *et al.*, 1984; Jeffries, 1985). The presence of monazite, xenotime and apatite as inclusions within biotite and the decrease in Y from monzogranites to leucogranites in the SMB is consistent with a model in which the LREE depletion is related to fractionation of such minerals (see more detailed discussion by Miller and Mittlefehldt, 1982, 1984; Mittlefehldt and Miller, 1983).

The leucogranite suites contrast with the monzogranites (also granodiorites, see Clarke and Muecke, 1981) in having distinctly concave patterns for the MREE-HREE. This is particularly noticeable in the group II leucogranites where the maxima in the chondrite-normalized patterns occur at Gd-Dy. Accompanying this marked change in the fractionation of the MREE and HREE is a flattening of the LREE profiles to $<10 \times$ chondrites. Although further depletion of the LREE may be accounted for by continued fractionation of accessory minerals, the similar Y contents of the group I and II

leucogranites suggest that an alternative mechanism, or combination of processes, may be required. The close proximity of group II leucogranites to pegmatites indicates that a fluid phase was exsolved from the melts and, thus, its effect on the REE patterns and abundances should be considered.

The ability of Cl to form stable complexes with LREE at magmatic temperatures provides a viable means to account for the type of REE patterns observed here. There is ample petrographic evidence indicating that aqueous fluids have interacted with the rocks, and the proximity of pegmatites indicates that a fluid phase did indeed exist. Thus, we consider the observed LREE depletion in group II leucogranites to be a result of fluid stripping, perhaps accompanying pegmatite development.

The concave patterns recorded in the REE profiles of group II leucogranites (Gd to Yb) may also be accounted for by the evolution of a fluid phase. These patterns are similar to published REE profiles for fluorite (Strong *et al.*, 1984; Clarke and Muecke, 1981; Chatterjee *et al.*, 1985b; Marchand *et al.*, 1976) and, therefore, the role of F complexing is considered a reasonable explanation. The peak of the REE profile at Gd-Dy suggests relatively dilute concentrations of F in the melt/fluid and that monofluoric complexes were dominant (Strong *et al.*, 1984). That F-bearing fluids were important in complexing the REE's is further demonstrated by a REE profile (Fig. 10) for a F-rich (6000 ppm) muscovite (analysis 36 in Tables 1 and 2) from Morley's pegmatite. The concave shape of the MREE-HREE is almost identical to that observed in the leucogranites. The enhanced Rb content (4840 ppm) of this muscovite also raises the possibility that some of the enrichment/depletion of this and other alkali elements in the leucogranites may be due to post-magmatic fluid-rock interaction.

We conclude, therefore, that on the basis of the REE distribution that there is an important

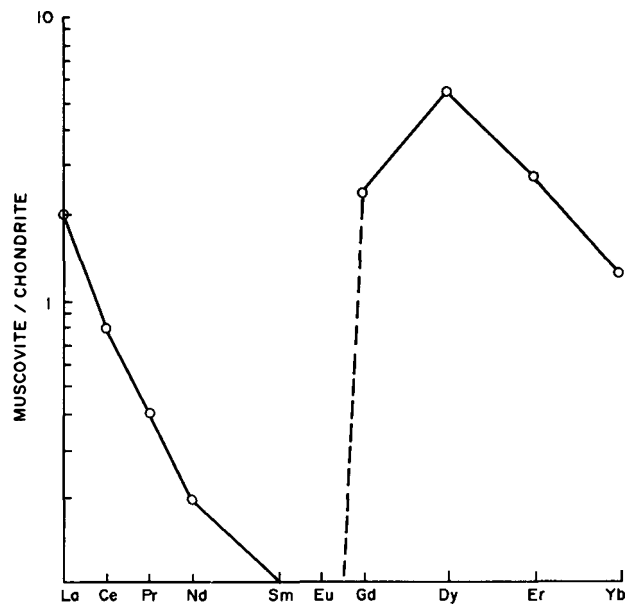


Fig. 10. Chondrite-normalized REE plot for muscovite separate from Morley's pegmatite.

transition from the progressive differentiation sequence granodiorite-monzogranite-leucogranite (I) which is dominated by silicate melt-crystal equilibria, to group II leucogranites. The latter appears to have involved, in addition to melt-crystal equilibria, an important component of aqueous fluid-phase equilibria. Although the role of a fluid phase is perhaps most easily recognized using the REE, the model calculations of Nabalek (1986) in a similar rock suite indicate that other trace element contents (e.g., Rb, Ba, Sr) would also be affected quite drastically. We note, therefore, that on log-log plots of Ba versus Rb and Sr the data for the leucogranites fall off the crystal fractionation trend defined by the data for the more mafic assemblages. Thus, because of the probable involvement of a fluid phase during the late-stage magmatic evolution of the SMB, we are hesitant in assigning a crystal-melt equilibria origin to many of the trace elements in the more evolved phases.

Source of Fluids and Post-Magmatic Disequilibrium

The use of oxygen isotopes permits one to identify the presence and affects of fluids of different derivations in igneous rocks (e.g., Taylor, 1978). The similarity of the whole-rock oxygen isotopic values for all the granitoids examined in this study, in addition to those of Longstaffe *et al.* (1980) and Chatterjee *et al.* (1985a), indicates at first sight that little modification of the primary oxygen isotopic signatures has occurred. In fact, the quartz-feldspar pairs and other mineral data of Longstaffe *et al.* (1980) indicate that oxygen isotopic equilibrium existed during magmatic crystallization and that the original isotopic character of these minerals has not been modified by post-magmatic processes. In contrast, the mineral data from a limited number of pegmatites and greisens suggest disequilibrium caused by late- to post-magmatic processes.

Discussing first the whole rock data, we note that there is no systematic variation of the $\delta^{18}\text{O}$ composition of the granites and that combining the data from all sources (i.e., Longstaffe *et al.*, 1981; Chatterjee *et al.*, 1985; this study) gives an average $\delta^{18}\text{O}$ composition of 10.77 ± 0.67 ‰ (N=54) for the SMB. The tight cluster and lack of any obvious relationship between $\delta^{18}\text{O}$ values and indices of differentiation indicates that the granitic magmas most likely evolved as a closed system (cf. Taylor, 1978). However, the observed range of 1.8 ‰ is in excess of that which can be related to crystal fractionation processes over such a small silica range (Taylor and Sheppard, 1986) and thus an alternative explanation is required. Because there is little compelling evidence to favour assimilation of Meguma Group lithologies ($\delta^{18}\text{O}$ of 11.6 ± 0.9 , N=31; Longstaffe *et al.*, 1980) as the cause of this variation (see Clarke and Halliday, 1980 for further discussion), we instead interpret the range in oxygen isotopic data as reflecting heterogeneity in the source region.

Focusing on the oxygen isotope data for mineral separates (Figs. 8, 9), we have noted that disequilibrium fractionations are recorded.

Because of the tendency of feldspar to re-equilibrate more readily than quartz at low temperatures, the shift of the data out of the magmatic field in Figure 9 is attributed to exchange of ^{18}O between an aqueous fluid and feldspar at low temperatures. The $\delta^{18}\text{O}$ composition of the fluids involved in this re-equilibration is estimated using mineral-water fractionations (Fig. 11). Although a range of 400–600°C is indicated for quartz in Figure 11, the higher temperature is considered more realistic and indicates a $\delta^{18}\text{O}$ of 9–10 ‰ (500–600°C) for the fluid. In contrast, the feldspars are interpreted to have exchanged with a fluid phase to lower temperatures based on their strongly perthitic textures and partial inversion to triclinic structures (unpublished X-ray diffraction data of Kontak). For temperatures of 400–500°C, the $\delta^{18}\text{O}$ of the fluid in equilibrium with feldspar is 7–9 ‰. Thus, the inferred range (7–10 ‰) of the $\delta^{18}\text{O}$ composition of the fluids based on both feldspar and quartz mineral-water fractionations is consistent with a magmatic reservoir, although a component of metamorphic-derived fluid cannot be excluded (Ohmoto, 1986).

In contrast, the $\delta^{18}\text{O}$ composition of the fluid responsible for greisen formation is markedly different (Fig. 11). For sample #28 a $\delta^{18}\text{O}$ value of 11.5–14 ‰ is calculated based on quartz-water fractionation for a temperature of 300–400°C. The enrichment of the fluid in ^{18}O indicates a certain component of either metamorphic-derived water or fluid which equilibrated isotopically with the nearby metasedimentary rocks. The feldspar in sample #29 appears to have retained more of its primary magmatic signature compared to quartz in sample #28, as the calculated $\delta^{18}\text{O}$ of the fluid is 7–10 ‰ for a similar temperature interval. The difference between these two greisens may be related to a much higher fluid-rock ratio in the former which resulted in complete conversion of the feldspar to a quartz-muscovite assemblage. Conversely, fluids with different oxygen isotopic signatures may have been involved.

CONCLUSIONS

A combined petrological and oxygen isotopic study of a suite of granitoid rocks in the SMB of Nova Scotia has revealed the following:

(1) The chemical evolution of the SMB is best accommodated by crystal-melt equilibrium processes in the early, less evolved members. However, in the more fractionated rocks, represented by leucogranites of the New Ross area spatially associated with pegmatites, chemical signatures (e.g., REE patterns) indicate that fluid-melt equilibria were also important.

(2) The LREE depletions observed in granites of the SMB are attributed to fractionation of accessory mineral phases, but fluid stripping was also an important process in the most evolved rocks. The concave profiles observed for the MREE-HREE part of chondrite-normalized diagrams in the most fractionated rocks are attributed to F-complexing via interaction of melts with a F-bearing aqueous fluid phase.

(3) The abundances of many trace elements, particularly the alkalis (Rb, Sr, Ba), in the fractionated suites are due to a combination of processes involving both magmatic and post-magmatic

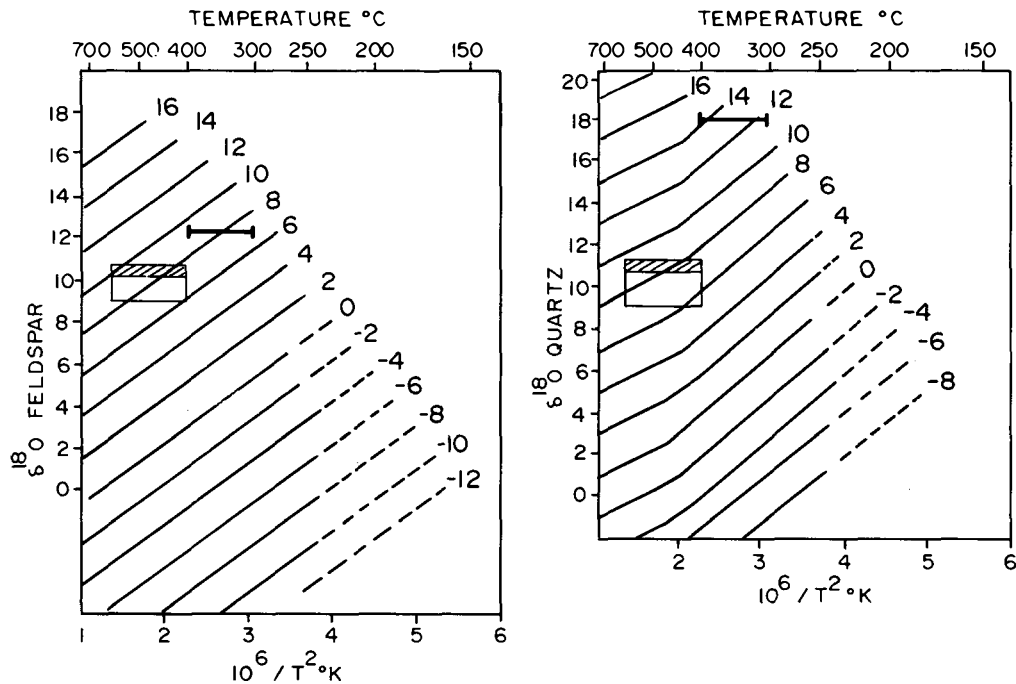


Fig. 11. $\delta^{18}\text{O}$ feldspar and quartz versus temperature plots showing oxygen-isotope data for feldspars and quartz. The temperature ranges are discussed in the text. The family of lines represents the $\delta^{18}\text{O}$ of H_2O in equilibrium with feldspar and quartz at given $\delta^{18}\text{O}$ for specific temperatures. Dashed lines indicate fractionations extrapolated to below the limits of equilibration. The upper part of the boxes contain most of the data and are the areas used for calculating the $\delta^{18}\text{O}$ values of the fluid discussed in the text. Calculations based on the fractionation factors (mineral-water) of Clayton *et al.* (1972) and O'Neil and Taylor (1967) for quartz and K-feldspar, respectively.

contributions.

(4) There is no measureable difference between the oxygen isotopic signatures of the major lithologic units of the SMB regardless of the degree of fractionation, proximity to country rock or presence of late-magmatic or early post-magmatic fluid interaction. The whole rock $\delta^{18}\text{O}$ values (9.5–11.4 ‰) are similar to those in other strongly peraluminous suites world wide and indicate a large component of crustal material in the genesis of the granites. The observed variation in $\delta^{18}\text{O}$ values is due to heterogeneity in the source region.

(5) Oxygen isotopic values of mineral separates from pegmatitic suites indicate that equilibrium fractionations were not maintained. This is attributed to low temperature (i.e., to 400°C) exchange of feldspars with cooling fluids of magmatic derivation. In contrast, isotopic signatures in greisens reflect interaction with a component of metamorphic-derived fluid.

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